RESEARCH IN LASER PROCESSES

A. V. Phelps

Joint Institute for Laboratory Astrophysics

Prepared for:

Office of Naval Research
Advanced Research Projects Agency

27 February 1975

090091

JOINT INSTITUTE FOR LABORATORY ASTROPHYSICS

UNIVERSITY OF COLORADO

NATIONAL BUREAU OF STANDARDS

AD A006986

SEMIANNUAL TECHNICAL REPORT

RESEARCH IN LASER PROCESSES

Research Sponsored by

Advanced Research Projects Agency

and

Office of Naval Research

ARPA Order No. 2683, Amendment 1
Program Code No. 4E20
Contractor: University of Colorado
Effective Date of Contract: July 1, 1974
Contract Expiration Date: June 30, 1975
Amount of Contract: $120,000

Contract No. N00014-67-A-0405-0008
Principal Investigator: A. V. Phelps
Telephone: 303-499-1000 Ext. 3604
Scientific Officer: Director,
Physics Program, ONR
Short Title of Work: Laser Processes

February 27, 1975

20

UNCLASSIFIED

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

REPORT DOCUMENTATION PAGE

READ INSTRUCTIONS BEFORE COMPLETING FORM

1. REPORT NUMBER: None

2. GOVT ACCESSION NO.:

3. RECIPIENT'S CATALOG NUMBER:

4. TITLE (and Subtitle): Semiannual Technical Report

5. TYPE OF REPORT & PERIOD COVERED: Interim 1 July 1974 to 31 Jan. 1975

6. PERFORMING ORG. REPORT NUMBER:

7. AUTHOR(s): A. V. Phelps

8. CONTRACT OR GRANT NUMBER(s): N00014-67-A-0405-0008

9. PERFORMING ORGANIZATION NAME AND ADDRESS:
University of Colorado
Joint Institute for Laboratory Astrophysics
Boulder, CO 80302

10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS:
NR 012-512/6-14-74, Code 421
ARPA Ord No 2683, Prog.Cd.4E20
ARPA Ord No 2683, Amd 1, Prog. Cd. 5E20

11. CONTROLLING OFFICE NAME AND ADDRESS:
Office of Naval Research
Department of the Navy
800 North Quincy Street, Arlington, VA 22217

12. REPORT DATE: 27 February 1975

13. NUMBER OF PAGES: 20

14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office):

15. SECURITY CLASS. (of this report): UNCLASSIFIED

15a. DECLASSIFICATION/DOWNGRADING SCHEDULE: (blank)

16. DISTRIBUTION STATEMENT (of this Report):

Distribution Unlimited

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report):

18. SUPPLEMENTARY NOTES:

19. KEY WORDS (Continue on reverse side if necessary and identify by block number):

Laser, Processes, Continuum radiation, Oxygen, CO_2 Lasers, CO Lasers

20. ABSTRACT (Continue on reverse side if necessary and identify by block number):

Measurements of the A-X band continuum radiation of the Ha-noble gas and Li-noble gas molecules have been used to deduce the gain for high-power laser operation. The Na-Xe, Li-He, and Li-He excimers look promising as high-power laser systems. An analysis of electron transport coefficients, excitation rate coefficients, and attachment and ionization coefficients for electrons in O_2 has been nearly completed. The contributions of various electron excitation processes to gas heating and thereby to arc formation in CO and CO_2 lasers have been evaluated.

DD FORM 1 JAN 73 1473 EDITION OF 1 NOV 65 IS OBSOLETE

PRICES SUBJECT TO CHANGE

UNCLASSIFIED

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

SEMIANNUAL TECHNICAL REPORT

This Semiannual Technical Report contains descriptions of work carried out under ONR Contract No. N0014-67-A-0405-008 and ARPA Order No. 2683, Amd. 1, and covers the period from 1 July 1974 to 31 January 1975. Section I is the Technical Report Summary while Sections II-V are more detailed descriptions of work carried out under the four projects supported by this contract.

I. TECHNICAL REPORT SUMMARY

The four projects being carried out in the area of Laser Processes under this contract are summarized below. More detailed discussions are given in Sect. II of this report.

1) Stability of Discharges in Weakly Ionized Gases.

The technical problem being investigated is the optimization of the use of electrical energy for the production of excited molecules in electrically excited gas lasers. In particular, the efficient use of electrical excitation in gas lasers requires that the discharge remain diffuse as the density of the excited molecules is raised as high as possible, i.e., that the discharge not form a constricted channel or arc. We are making numerical calculations of the development of pulsed discharges in high pressure gas mixtures of interest for high power lasers, e.g., CO_2 and CO lasers. Although our calculations thus far are limited to radial flow, they show that for a wide range of gas pressures, electric fields and time scales the most important process leading to the eventual constriction of the discharge is the flow of heated gas and the resultant increase in the ratio of the electric field to the gas density and in the local rate of ionization of the gas. The next step in the modeling is to allow for axial propagation of the discharge as is often observed experimentally.

2) Electron Transport and Ionization Coefficients

The objective of this project is to supply accurate electron transport coefficients, electron excitation and ionization coefficients, electron collision cross sections, and excited state relaxation data for use in the electric discharge models discussed above and being carried out in laser

development laboratories. The procedures used include the assessment of the accuracy to be expected from various experimental methods for the measurement of electron transport coefficients; the analysis of published and unpublished electron transport data and collision cross section data to obtain the best available set of electron cross section data, e.g., a set which is consistent with the limited amount of measured electron transport coefficients in gases of laser interest; and the compilation and application to laser gas mixtures of data on the collisional and radiative destruction of electronically excited molecules. Specific results include the generation of an improved set of electron excitation cross sections and rate coefficients for oxygen and the improvement of estimates of the heating of gases such as N_2 and CO as the result of the excitation of rotational and electronic states. This work will be extended through the application of recently developed techniques to the measurement of electron excitation rate coefficients for slowly radiating (metastable) states of molecules such as oxygen.

3) Generation and Interpretation of Molecular Continuum Radiation

The technical problem addressed by this project is the evaluation of usefulness of a number of high pressure, metal atom-rare gas atom mixtures as the active laser media for high power lasers at wavelengths in the near ultraviolet, the visible and the near infrared. The approach is to measure the wavelength dependent fluorescence when the mixture is irradiated with radiation which is readily absorbed by the metal atoms, i.e., the resonance radiation characteristic of the metal. This pressure and/or temperature dependent data is then used to a) predict the temperature dependent coefficients for stimulated emission and absorption which are needed for the assessment of the utility of the mixture for laser purposes

and b) construct potential energy curves for the molecule made up of the metal vapor and rare gas atoms so as to provide a basis for the prediction of the usefulness of mixtures not yet studied experimentally. Recent results, discussed in Sect. IV show that, as we predicted from theory several years ago, the mixture of Li and Xe does offer the possibility of reasonable gain at wavelengths near 9000Å. Another potentially useful combination is the mixture of Li and He.

4) Scattering and Transport of Resonance Radiation in Gases

The technical problem considered here is the accurate and rapid prediction of the experimentally important loss of excited atoms from laser plasmas through the transport of resonance radiation to the walls of the laser. The method used is the experimental measurement of the magnitude and wavelength dependence of the backscattering of radiation incident on a cell filled with metal vapor, e.g., sodium or potassium. The experimentally measured spectral intensity is then compared with theoretical predictions so as to evaluate the accuracy of the theory. We are particularly concerned with developing and testing simple theories of resonance radiation transport which can then be fitted into the complex models of laser discharges, etc.

II. STABILITY OF DISCHARGES IN WEAKLY IONIZED GASES (Drs. E. F. Jaeger - to 10/74 - and A. V. Phelps)

The numerical model previously used to discribe the growth of current in pulsed electrical discharges in helium and in CO_2-N_2-He laser mixtures has been successfully extended to describe the role of vibrational relaxation. This extension required the addition of a partial differential equation to represent the energy content of each mode of the vibrationally excited molecules. This equation, along with the equations required by the inclusion of negative ions, results in a set of at least nine coupled partial differential equations. Because of the large amount of computer time required only a few sets of parameters have been investigated and only one mode (the coupled 001 mode of CO_2 and the vibrational excitation of N_2) is included.

Figure 1 shows the results of a calculation carried out for a 1:2:3 mixture of CO_2:N_2:He for conditions which lead to an average current density in the diffuse discharge of 100 A/cm^2. The curves for the radial distribution of the translational-rotational temperature, T_{TR}, and for the vibrational temperature, T_V, show that the two temperatures are very nearly equal at the center of the discharge where the translational temperature is high and that the temperatures are widely different near the edge of the discharge where the translational temperature is low. Obviously this frame is for times beyond that at which the gas would provide useful gain. Note the structure in the electron density n_e, fractional gas density change $\Delta N/N$ and gas velocity v_N profiles.

Figure 2 shows calculated electron density and temperature transients for the same CO_2 laser mixture but for conditions corresponding to an

average discharge current of 1 A/cm^2. As expected there is a less rapid build up of the translational-rotational temperature of the gas and of the electron density when vibrational relaxation is suppressed. Again we see the rapid approach of the vibrational temperature T_V to the translational temperature T_{TR} when the translational temperatures exceeds about 600 K.

A paper describing the first phase of our theoretical studies of the growth of constricted discharges in laser gases has been submitted for publication. The paper is entitled "Growth of Thermal Constrictions in a Weakly Ionized Gas Discharge: Basic Model and Numerical Solutions for Helium" by E. F. Jaeger, L. Oster and A. V. Phelps.

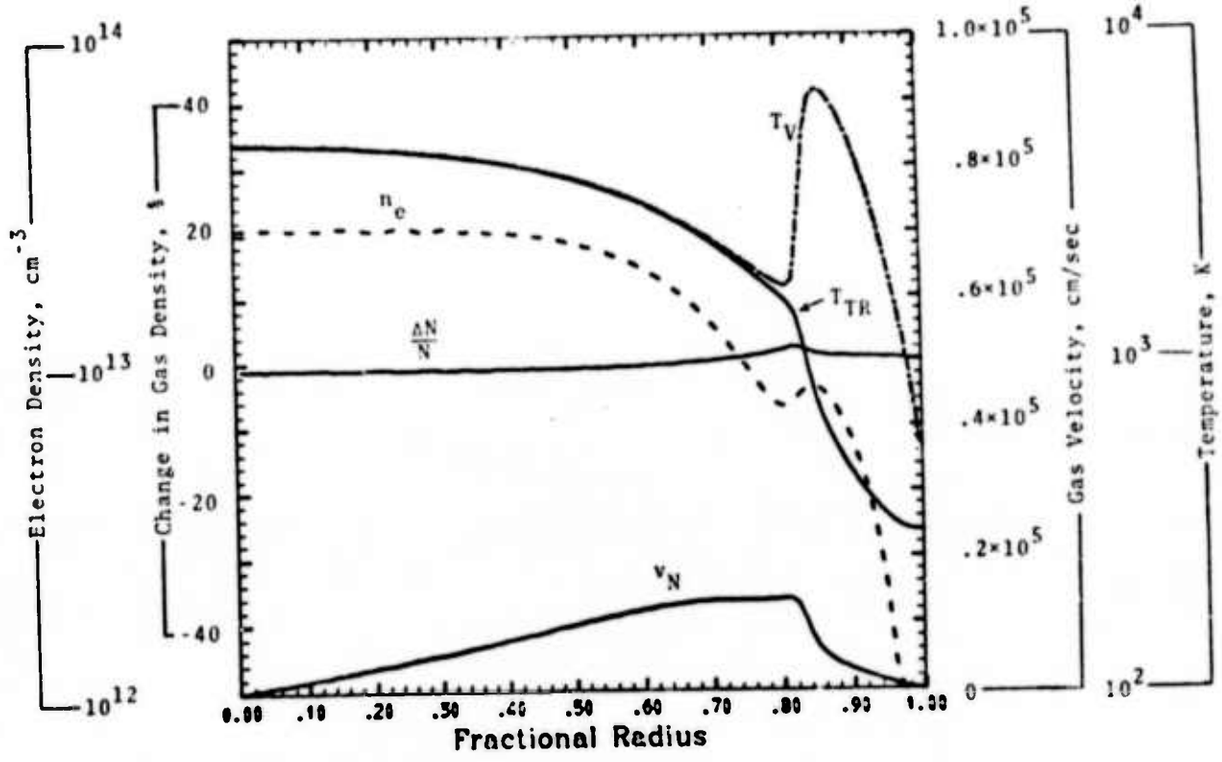

Fig. 1. Radial variation of translational-rotational temperature T_{TR}, vibrational temperature T_V, electron density n_e, fractional change in gas density $\Delta N/N$, and radial gas velocity v_N for 1:2:3 mixture of CO_2:N_2:He at 1000 torr at 7 μs after application of 18 kV/cm in a 20 cm dia tube with an external resistance of 0.4 ohm per cm of discharge length. The average discharge current is approximately 100 A/cm^2.

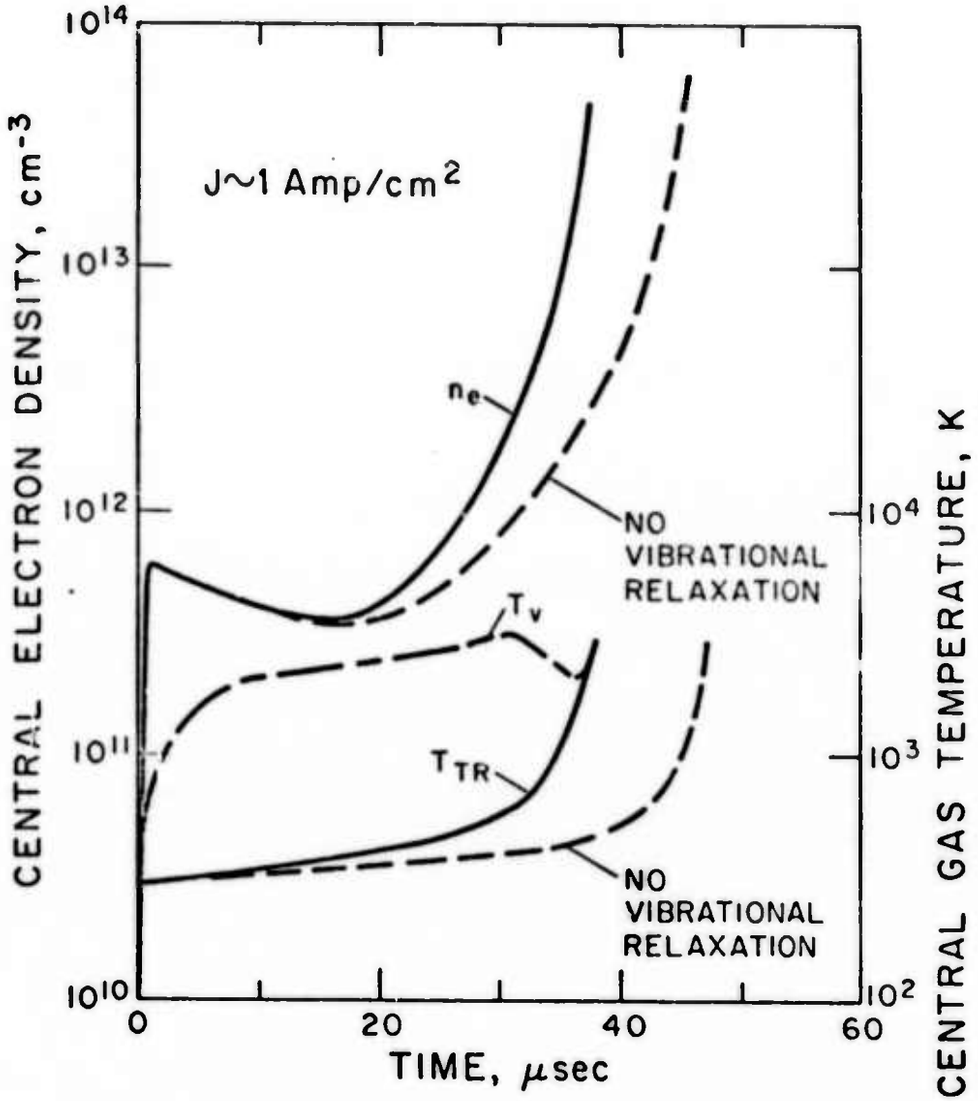

Fig. 2. Temporal variation of electron density n_e. vibrational temperature T_V, and translational-rotational temperature T_{TR} for the same conditions as Fig. 1 except that the external resistance has been increased to 50 ohm per cm of discharge so that the average current prior to 30 μs is about 1 A/cm^2. The short dashed curve shows the time variation of n_e and T_{TR} when vibrational relaxation of the CO_2(001) and N_2 is reduced to negligible values.

III. ELECTRON TRANSPORT AND IONIZATION COEFFICIENTS (Drs. J. H. Whealton and A. V. Phelps)

1) An analyses of electron transport coefficients, excitation rate coefficients and attachment and ionization coefficients for electrons in O_2 has been nearly completed. This analysis represents a significant improvement over our previous (1964) work in that where possible it incorporates the results of recent electron beam studies of 3-body attachment, vibrational excitation, metastable excitation and total excitation. Although there is no experimental electron excitation rate coefficient data for vibrational excitation, our analysis shows that for mean electron energies between 0.2 and about 1.5 eV either the published vibrational excitation cross sections are a factor of two too small or that the energy loss to non-resonant rotational excitation is larger than energy loss to vibrational excitation. While we prefer the former explanation, an experimental measurement will be required to settle the question. It is to be noted that in recent months we have received requests from four different DoD and industrial laboratories for improved electron excitation data for O_2. One laser system being considered is that involving excitation transfer from $O_2(a^1\Delta_g)$ molecules to the upper laser level of the iodine atom.

2) The results of an evaluation of collisions processes leading to gas heating and thereby contributing to arc formation were presented to the "CO Glow Collapse Working Group" at AFWL on 16 September. The processes considered were a) the excitation of rotational states of molecules by electrons with the subsequent rotational relaxation and b) the relaxation of electronically excited states of the molecules. Unfortunately, the

only estimates of the cross sections for pure rotational excitation by electrons at the energies of interest are based on untested theory of excitation via resonance states and apply only to N_2. We have adapted these theoretical cross sections to our computer codes for calculating electron transport coefficients in gas laser mixtures and find that the contribution of this process to heating of the gas varies from about 5% to about 1.3% as the electron mean energy is raised from 1 to 2.5 eV. The theoretical estimates of the heating by rotational relaxation as the result of simultaneous vibrational and rotational excitation have not yet been adapted to our computer code but are expected to approximately double the gas heating rate. At present we can only speculate that the contribution of rotational excitation to the heating of CO will be about the same as for N_2. Fortunately, our investigation has led to the initiation of an experimental measurement of rotational excitation by Schulz and coworkers at Yale. The available data on the excitation and relaxation of electronically excited states of N_2 shown in Table I suggests that it is possible to store significant amounts of excitation energy ($\sim$10%) for times of the order of 20 μs with typical CO_2-N_2-He laser discharge mixtures at atmospheric pressure. As shown by Table II the storage times would be much shorter for CO at comparable CO pressures. Of course, electronic excitation is relatively unimportant for most e-beam sustained lasers.

Table I. Gas Heating by Excited State Quenching in 1:2:3 of CO_2:N_2:He for E/N = 4.75×10^{-16} V-cm^2

% Energy into Level(s)	Energy Loss eV	State(s)	A value sec^{-1}	Quenching Rate Coef. cm^3/sec	Lifetime·1 atm μ sec	E_{max}, eV, Q_{max} cm^2
0.7	0.02	N_2 rot				2.2,2.8×10^{-16}
18.5	.08→2.5	$CO_2(X^1\Sigma_g^+, v>0)$				3.4,5×10^{-16}
51.0	.3→2.4	$N_2(X^1\Sigma_g^+, v>0)$				2.2,5×10^{-16}
6.5	6.2	$N_2(A^3\Sigma_u^+)$	0.5(A→X)	$10^{-14}[CO_2]+6\times10^{-14}[H_2O]$ $+2\times10^{-9}[e] + 10^{-10}\ [N_2(A)]$	>24	10.5,2.8×10^{-17}
0.4	7.2	$N_2(W^3\Delta_u)$	$\sim10^5$(W⇄A)		<10(v>1)	20,8×10^{-18}
10.2	7.4	$N_2(B^3\pi_g)$	10^5(B→A)	$2\times10^{-12}[N_2]$	0.05	10.5,6×10^{-17}
2.2	8.4	$N_2(a^1\pi_g)$	7×10^3(a→X)			18,4×10^{-18}
0.7	11.2	$N_2(C^3\pi_u)$	3×10^7(C→B)	$10^{-11}[N_2]$	0.01	14,4×10^{-17}
		$N_2(E^3\Sigma_g^+)$		$\sim10^{-10}[N_2]$	0.001	
0.09	12.4	N_2(?)				90,1.5×10^{-16}
0.03	14.4	N_2(?)				90,1.8×10^{-16}
0.01	15.5	N_2^+	0			100,2.5×10^{-16}
6.4	7	CO_2(?)				8,6×10^{-17}
1.7	10.5	CO_2(?)				100,6×10^{-17}

Table II. Gas Heating by Excited State Quenching in CO

% Energy	Threshold eV	State(s)	A Value sec^{-1}	Quenching Rate Coef. cm^3/sec	Lifetime·1 atm μ sec	E_{max}, Q_{max} eV, cm^2
	0.3-3 eV	$CO(X^1\Sigma^+)$				
	6.1	$CO(a^3\pi)$	1.4×10^2	$1.4\times10^{-12}[CO]+10^{-11}[N_2]$	<0.1	$10,1.1\times10^{-16}$
	7.9	$CO(A^1\pi)$	10^8	$10^{-12}[He]+3\times10^{-11}[Ar]$	<0.02	$23,5\times10^{-17}$
	9.5	$CO(D^1\Delta?)$	10^4			$15.5,3\times10^{-18}$
	10.4	$CO(b^3\Sigma^+)$	1.6×10^7			$12,8\times10^{-18}$
	10.7	$CO(C^1\Sigma^+)$				$30,1.7\times10^{-17}$
	10.8	$CO(E^1\pi)$				$30,1.1\times10^{-17}$
	11.4	$CO(B^1\Sigma^+)$	10^8	$5\times10^{-11}[CO]$	<0.02	$30,2.6\times10^{-18}$
	12.1	$CO(C^1\Sigma^+)$				$30,1.1\times10^{-17}$
		$CO(E^1\pi)$				$30,7\times10^{-18}$
	13.5	CO(?)				$45,3.5\times10^{-17}$
	14.0	CO^+				$100,3\times10^{-16}$

IV. GENERATION AND INTERPRETATION OF MOLECULAR CONTINUUM RADIATION (Drs. A. C. Gallagher and R. Scheps)

1) We have measured the A-X band continuum radiation of the Li-noble gas molecules, inferring potentials, excimer formation rates, and laser gain for possible high-power laser use. The Li-He and Li-Xe excimers look particularly attractive since their emission extends a wavelength beyond the Li_2 absorption. Examples of these results are the measurements of the normalized fluorescent intensity shown in Figs. 3 and 4 for the Li-He and Li-Xe systems. These results are used to calculate intermolecular potentials for the LiHe and LiXe molecules (not shown) and to calculate the absorption and stimulated emission coefficients shown in Figs. 5 and 6 for the Li-He and Li-Xe systems under the stated conditions of temperature, Li vapor density, rare gas density and fractional Li excitation. The interfering effects of absorption by Li_2 molecules at densities appropriate to these conditions are indicated by the dashed lines. The curves for Li_2 are smoothed curves representing the average effects of numerous bands.

2) We have modeled the gain behavior of the A-X band of the alkali-domer molecules (Li_2, Na_2, etc.) for conditions that apply in a high-power laser system. These systems appear to warrant serious consideration as high-power laser candidates for the 700-1200 nm wavelength region. Details of these calculations are given in JILA Report No. 114 of October 15, 1974 by G. York and A. C. Gallagher.

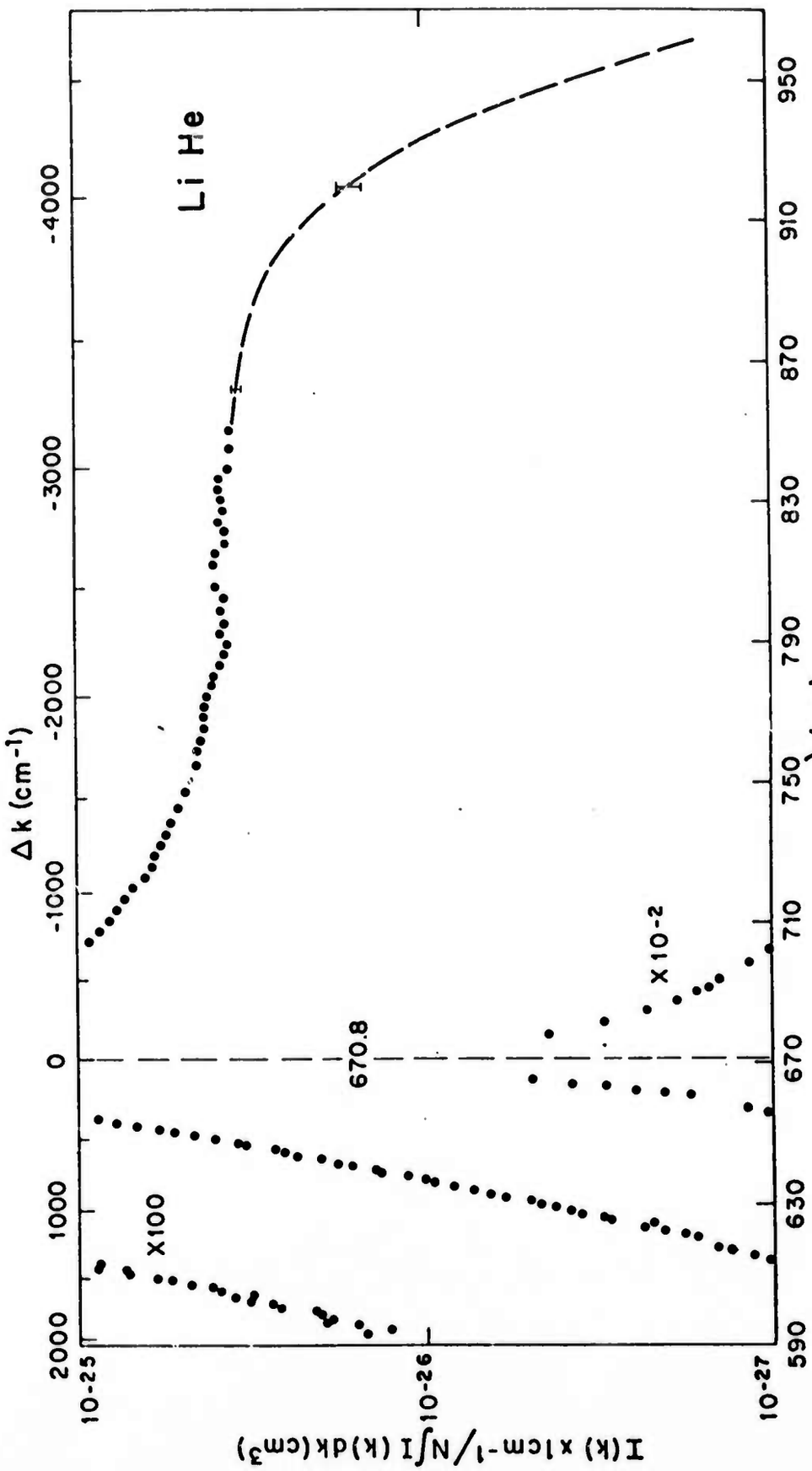

Fig. 3. The emission coefficient of Li-He as a function of λ. The He pressure is 600 torr, while 40 torr data (not indicated) were indistinguishable at ~3% accuracy. The spectrometer resolution is 15A and no corrections for this width are included or deemed necessary.

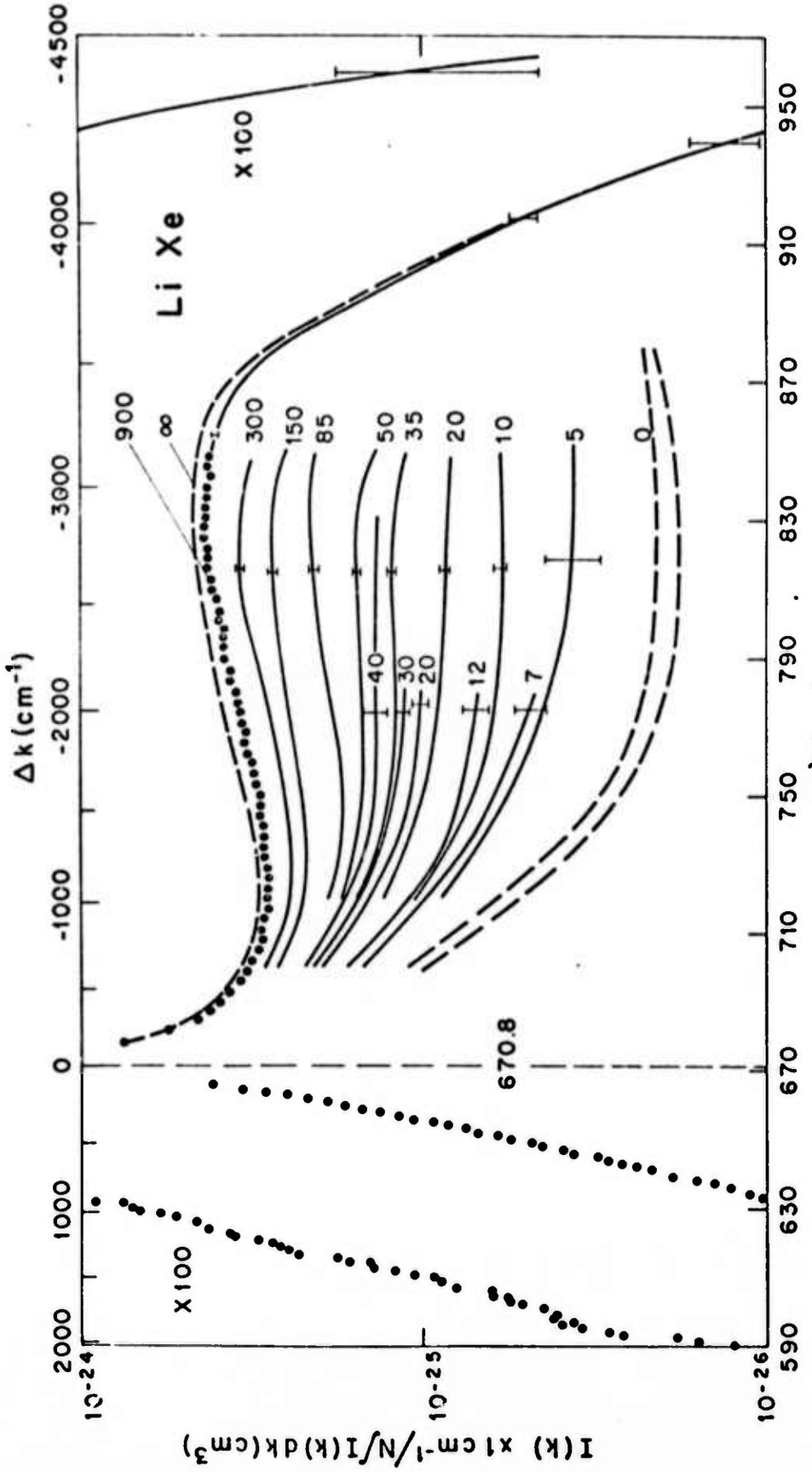

Fig. 4. Same as Fig. 3 for Li-Xe. At $\lambda > 860$ nm, only high pressure data were taken since a less efficient photomultiplier had to be used. The zero pressure limits are extrapolated beyond 860 nm for use in estimating the potentials in the corresponding radial region.

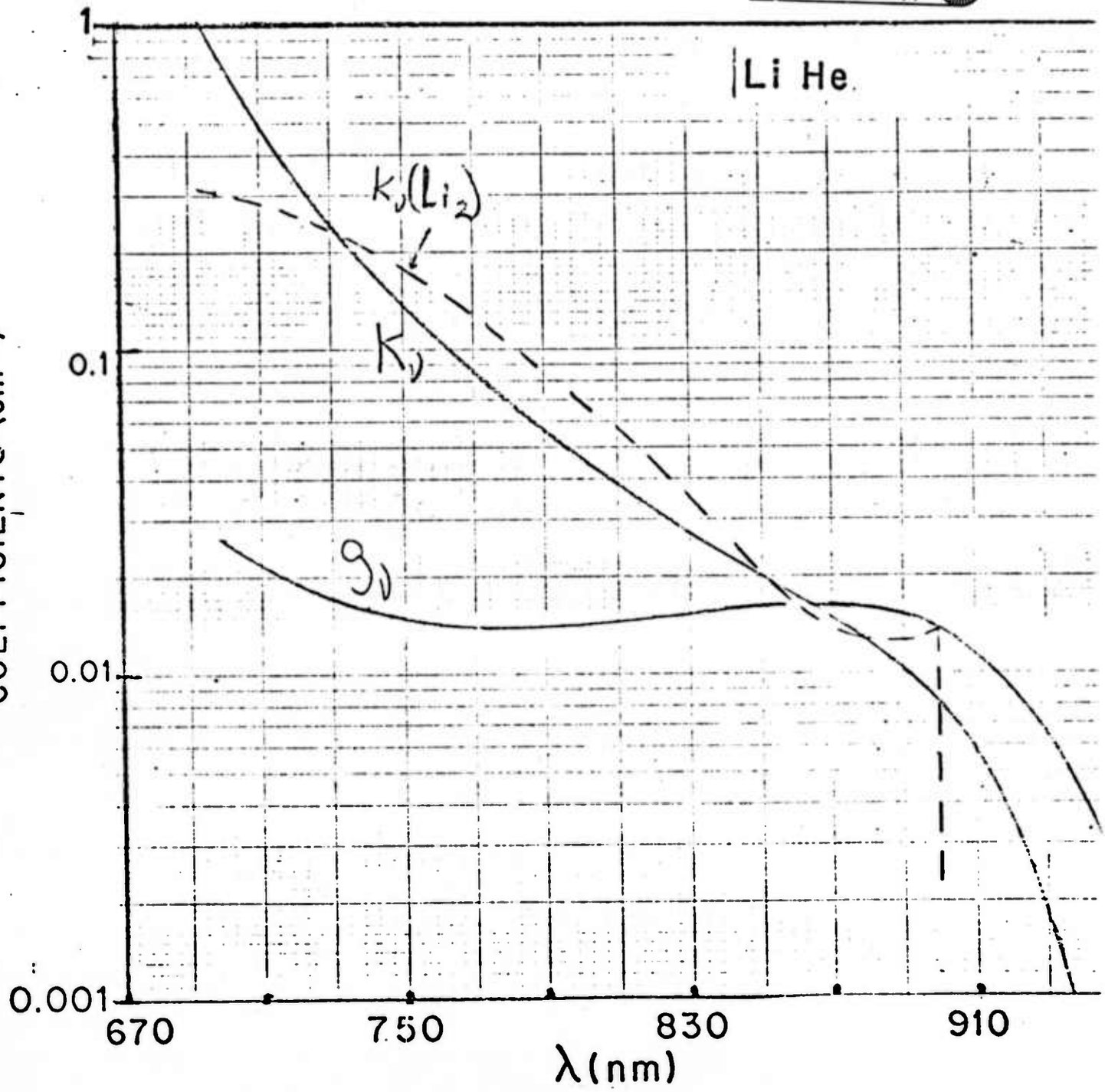

Fig. 5 Lower limits for the absorption K_ν and stimulated emission g_ν coefficients for Li-He vapor at 1180°K. Concentrations [He] = $2 \times 10^{20}/cm^3$, [Li] = $10^{17}/cm^3$, and [Li*] = 0.05 [Li] are assumed. Absorption by Li_2 is also indicated.

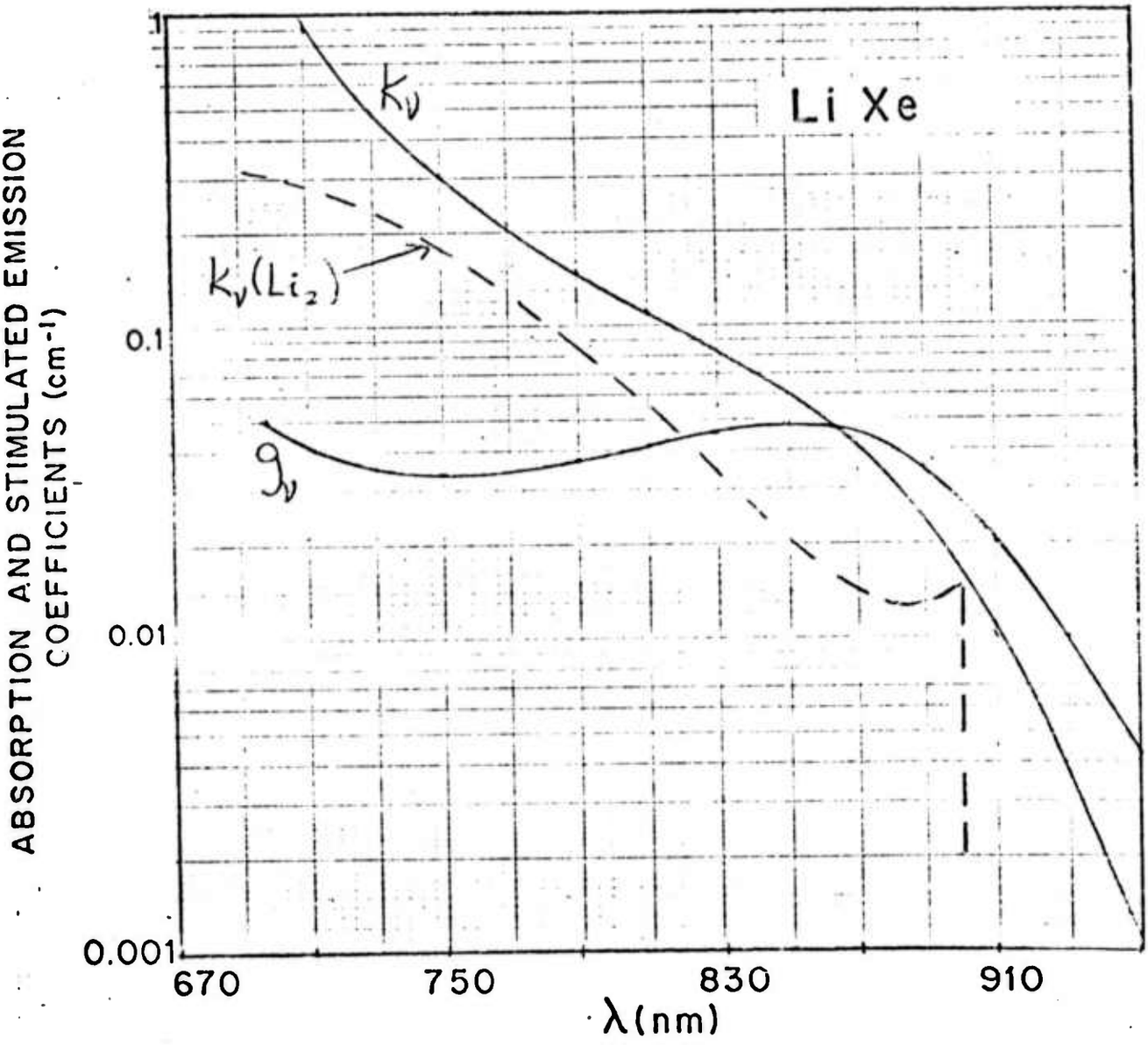

Fig. 6. Absorption K_ν and stimulated emission g_ν coefficients for Li-Xe vapor at 1180°K. Concentrations of [Xe] = $2 \times 10^{20}/cm^3$, [Li] = $10^{17}/cm^3$, and [Li*] = 0.5 [Li] are assumed. Absorption by Li_2 is also indicated.

V. SCATTERING AND TRANSPORT OF RESONANCE RADIATION IN GASES (Drs. T. Fujimoto and A. V. Phelps)

The development of experimental and theoretical techniques for the prediction of the transport of non-coherent radiation, e.g., resonance radiation. under conditions of large self absorption continues. The experimental measurements to be made have been reduced to those of the variation with wavelength of the fractional diffuse backscattering and of the absorption of "white" light incident on the alkali metal vapor filled cell. This reduction considerably simplifies the data recording and improves the odds of satisfactory operation of all the various components of the experiment. Preliminary measurements of absorption profiles using a previously fabricated cell show good agreement between measured and calculated values of the total absorption. Unfortunately, the cell windows have become too coated with material from the vacuum seals to allow satisfactory backscattering measurements.

We recently have purchased new cell windows with vacuum seals which are claimed by the manufacturer to be much more resistant to alkali metal vapors than the vacuum seals previously available. We are currently attempting to fabricate an absorption cell using these windows. The very detailed theory of the transport of resonance radiation developed earlier for us by D. Hummer and P. Kunasz is being readied for application to our measurements diffuse backscattering in Na and K vapors. In addition to radiation transport, this theory includes the effects of collisional broadening of the resonance line, of collisional mixing and quenching of the excited atoms, and of the diffusion of excited atoms to the cell windows.